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I, TERESA KOLODZIEJCZYK, TEAM LEADER EXAMINATION SUPPORT AND SALES hereby certify that annexed is a true copy of the Provisional specification in connection with Application No. PS 1464 for a patent by COMPCO PTY LTD as filed on 28 March 2002.

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Ninth day of April 2003

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PROVISIONAL SPECIFICATION

Applicant(s):

COMPCO PTY LTD

A.C.N. 070 193 805

Invention Title:

NANOFILLER COMPOSITIONS

The invention is described in the following statement:

NANOFILLER COMPOSITIONS

FIELD OF THE INVENTION

The present invention relates to nanofiller compositions, processes for their preparation and articles composed of them, in particular nanofiller compositions containing cross-linkable and/or cross-linked olefin (co)polymers such as polyethylene. These nanofiller compositions possess advantageous properties, more specifically, increased barrier properties, strength and higher heat distortion temperatures which makes them useful in various applications including medical, automotive, electrical, construction and food applications.

BACKGROUND OF THE INVENTION

Thermoplastic polymers such as thermoplastic polypropylene have been mixed with fillers such as clays or calcium carbonate to produce compositions which only show minimal improvement in mechanical and chemical properties with deterioration during processing.

When nanofillers were added to thermoplastic polymers such as polypropylene in reduced amounts compared to standard fillers, some improvements in properties were obtained such as increased mechanical properties including stress crack resistance and tensile strength, reduction in gas or liquid permeability and increases in crystalline melting temperatures and flame retardancy. However, despite the addition of nanofillers, the polypropylene is still thermoplastic and its thermo-mechanical properties, tensile strength, resistance to permeability of gases or liquids, resistance to swelling and solvents and flame retardance at higher temperatures including in heat and sunshine is still reduced. The Stress Crack Resistance (SCR) and Environmental Stress Crack Resistance (ESCR) of most thermoplastics at greater than ambient temperatures such as in cars and cables is weakened, insufficient and

fails both in tests and use.

The swellability and solubility of thermoplastics in certain solvents, fuels, oils, chemicals strongly increases at elevated temperatures up to unacceptable limits and they may dissolve at elevated temperatures or when boiled or extracted in solvents at higher temperatures. Swellability means deterioration in properties, softening, increase in dimensions, mechanical weakening to the point of structural failure of the product made therefrom and ultimately, in some cases to dissolution of the product.

The flame retardance, in case of a test or in a real fire, can be reduced or impaired by the dripping thermoplastic polymer in particular in the flame temperature ranges. Dripping can result in acceleration of the fire due to hot, molten, even burning drops of polymer falling on other parts of products under or in the vicinity of the burning polymer.

Thus, the improvements observed by the addition of nanofillers to thermoplastic polymers were not and are not sufficient to reach the higher levels of performance required for increased safety levels of the products made therefrom both mechanically and thermo-mechanically, in particular at higher temperatures or in other difficult conditions such as exposure to chemicals, solvents, oils, fuels or short circuits. These properties are very important for products such as fuel tanks for automobiles, containers for solvents, chemicals, cables, aerial cables, power cables, foils and films. Furthermore, such compositions cannot be used to make heat shrinkable products for joints, sleeves, tubes, pipes, films and packaging.

A requirement accordingly exists for a nanofiller composition containing thermoplastic polymers which has improved properties so that the products made from these compositions perform well , particularly at temperatures above ambient and/or in difficult environments such as

exposure to chemicals, solvents, oils, fuels or short circuits.

SUMMARY OF THE INVENTION

The present invention provides a nanofiller composition which comprises a cross-linkable and/or cross-linked olefin (co)polymer and an intercalated nanofiller.

The present invention also provides a process for preparing a nanofiller composition which comprises either:

(a) mixing and exfoliating and/or delaminating in one step a cross-linkable and/or cross-linked olefin (co)polymer and an intercalated nanofiller;

(b) mixing a cross-linkable and/or cross-linked olefin (co)polymer with an intercalated nanofiller; and

exfoliating and/or delaminating at least part of the nanofiller; or

(c) exfoliating and/or delaminating at least part of an intercalated nanofiller; and

mixing the exfoliated and/or delaminated intercalated nanofiller with a cross-linkable and/or cross-linked olefin (co)polymer.

In another aspect of the process, the olefin (co)polymer and/or nanofiller are subjected to grafting either before, during or after the mixing and exfoliating and/or delaminating step(s). The grafting involves treating the olefin (co)polymer and/or nanofiller with an organic silane which is then grafted onto the (co)polymer and/or intercalated into the nanofiller using a free radical initiator.

The present invention further provides an article which is wholly or partly composed of the nanofiller composition defined above.

In a further aspect, the present invention provides a process for preparing the article defined above which comprises either:

(a) forming or shaping the nanofiller composition defined above;

(b) combining at least one layer of the nanofiller composition with at least one other layer; or

(c) heating and stretching the nanofiller composition defined above and cooling the stretched composition.

DETAILED DESCRIPTION OF THE INVENTION

Suitable olefin (co)polymers include ethylene (co)polymers such as polyethylene and ethylene based alkene or alphaolefin copolymers, for example, high density polyethylene (HDPE), medium density polyethylene (MDPE), linear low density polyethylene (LLDPE), low density polyethylene (LDPE), very low density polyethylene (VLDPE), and ultra low density polyethylene (ULDPE) ; ethylene hexene copolymers and ethylene octene copolymers; butylene (co)polymers such as polybutylene and polyisobutylene; ethylene-propylene copolymers (EPM); ethylene-propylene-diene terpolymers (EPDM); ethylene-butylene copolymers (EBM) and terpolymers (EBDM); ethylene-vinylsilane (co)polymers; terpolymers of ethylene with ethylene acrylate and acrylic acid (EAA) or methacrylic acid (EMA); and copolymers of ethylene with ethylacrylate (EEA), butyl-acrylate (EBA) or vinyl acetate (EVA). It will be appreciated that these olefin (co)polymers may also be in the form of metallocene catalyst (co)polymers.

The olefin (co)polymers or part of the olefin (co)polymers may be grafted with carboxylic acid groups such as maleic anhydride or acid or fumaric anhydride or acid which may facilitate the exfoliation and/or delamination of the nanofiller. Examples of grafted olefin (co)polymers suitable for use in the present invention include maleic anhydride (MAH) or maleic acid grafted copolymers such as LDPE-MAH, HDPE-MAH, EP-MAH, EPR-MAH, PE-MAH or PP-MAH.

The ethylene content of the ethylene-propylene copolymers is preferably about 10 to about 99.9% by

weight, more preferably about 40 to about 99.9% by weight, most preferably about 75 to about 99.9% by weight. It will be understood that the term "% by weight" as used herein is based on the total weight of (co)polymer.

The vinyl acetate content of the ethylene-vinyl acetate copolymer (EVA) is about 3 to about 80%, preferably about 9 to about 70% by weight, more preferably about 30 to about 50% by weight. The olefin (co)polymer may be an elastomer or a plastomer. Elastomers and plastomers can be characterised by means of specific gravity (S.G.) and other properties such as the differential scanning calorimetry (DSC) melting peak, Shore A hardness and elasticity modulus. Such properties will vary depending on the type of olefin (co)polymer and its method of manufacture and the amount of (co)monomer present. However, generally elastomers are elastomeric and plastomers are plastomeric or thermoplastic elastomeric and flexible.

Preferably, at least about 40% to about 50% by weight, more preferably at least about 60% by weight of the total (co)polymer is a plastomer with the balance being an elastomer. Examples of plastomers include polyethylene such as HDPE, MDPE, LDPE, LLDPE or VLDPE; EVA with up to about 28% vinyl acetate; EPM with up to about 25% propylene; and ethylene octane copolymers with S.G. up to about 0.886. The elastomers include ethylene octane copolymers with an S.G. of at least about 0.887; ULDPE; ethylene propylene copolymers such as terpolymers with propylene co-monomers of greater than about 30%; ethylene vinyl acetate copolymers with greater than about 30% vinyl acetate; EPDM; EPM; and EPR.

The term "cross-linkable and/or cross-linked" is used herein in its broadest sense and refers to the olefin (co)polymer being cross-linked or at least capable of being cross-linked at a later stage. It will be understood that at least one olefin (co)polymer in the composition may be cross-linkable and/or cross-linked and

such a (co)polymer preferably forms at least about 40% by weight of the total (co)polymer component.

The term "nanofiller" is used herein in its broadest sense and refers to fillers having a particle size in the nanometre (nm) range, preferably about 1 to about 100nm. The thickness of the particles is about 1 nm to about 100 nm and the diameter or length or width can be up to about 500 nm. When the nanofillers are exfoliated, their thickness is reduced to about 1 to about 3 nm.

The term "intercalated" is used herein in its broadest sense and refers to a platelet-like or layered structure. The layers of the nanofiller which are generally composed of silicate are intercalated with ionic or polar substances including quaternary ammonium salts, such as, optionally substituted long chain hydrocarbon quaternary ammonium salts, for example, benzyl or alkyl substituted long chain hydrocarbon quaternary ammonium salts, alkyl substituted tallow or hydrogenated tallow quaternary ammonium salts; or bis-hydroxyethyl quaternary ammonium salts. Suitable counter anions for the quaternary ammonium cations include halides such as chloride or methyl sulphate.

The intercalated nanofiller may be an intercalated mineral nanofiller or clay which is either synthetic or natural, such as, montmorillonite, bentonite, smectite and phyllosilicate which have been intercalated by organic modification with the ionic or polar substances described above and may be sold under the trade names Cloisite, Nanofil, Tixogel and Kunipia.

Nanofillers such as montmorillonite have an anisotropic, plate like, high aspect-ratio morphology which leads to a long and tortuous diffusion path through the structure and an improved barrier to permeation, particularly when used in combination with the cross-linked olefin (co)polymers of the present invention.

The amount of nanofiller is generally about 0.1 to about 10%, preferably about 1 to about 6%, more

preferably about is 3 to about 5% by weight.

It will be appreciated that known fillers may also be included in the composition. Suitable known fillers include inorganic and/or mineral fillers such as clays which may be calcined; talc; mica; kaolin; alkaline earth metal carbonates, for example, calcium carbonate, magnesium calcium carbonate or hydrated basic magnesium carbonate; and metal hydroxides, for example, aluminum or magnesium hydroxide. The fillers may optionally be coated with, for example, stearic acid, stearates such as calcium stearate, silanes such as vinyl silane, siloxanes and/or titanates. While such coatings can be used to coat the fillers, they can also be added simultaneously, sequentially and/or separately with the fillers.

The olefin (co)polymer and/or nanofiller can be grafted using an organic silane and a free radical initiator. In a preferred embodiment, effective amounts of organic silane and peroxide are added to the (co)polymer and/or nanofiller either before or during the mixing step and then grafted onto the copolymer at temperatures preferably of about 150°C to about 240°C and more preferably about 180°C to about 230°C. In a particularly preferred embodiment, the silane and the peroxide are added to both the (co)polymer and/or nanofiller in a one step procedure which facilitates exfoliation and/or delamination of the nanofiller. In an alternative embodiment, the (co)polymer is grafted using the organic silane and peroxide and then mixed with the nanofiller followed by exfoliation and/or delamination.

Suitable organic silanes include vinyl silanes, for example, vinyl alkoxy silane such as vinyl-tris-methoxy-silane, vinyl-tris-methoxy-ethoxy-silane, vinyl-tris-ethoxy-silane, vinyl-methyl-dimethoxy-silane and gama-methacryl-oxypropyl-tris-methoxy-silane; amino silanes, for example amino propyl triethoxy silane (silane A110) or amino propyl trimethoxy silane (Silane A-1120); or long aliphatic hydrocarbon chain silanes. The vinyl

silane is preferably added in an amount from about 0.5 to about 2% by weight of the (co)polymer, more preferably about 0.8 to about 1.6% by weight.

The term "free radical initiator" is used herein in its broadest sense and refers to an unstable molecule or compound which generates free radicals. Examples of suitable initiators include peroxides such as dicumyl peroxide, di-tertiary-butyl peroxide, di-tertiary-butyl-cumyl peroxide and bis-tertiary-butyl-cumyl peroxide i.e., Perkadox 14. The free radical initiator is preferably added in an amount of about 0.1 to about 0.2% by weight calculated on the amount of (co)polymer, more preferably about 0.15 to about 0.2% by weight.

The (co)polymers, compositions and/or articles of the present invention may be cross-linked by adding cross-linking catalysts such as organic peroxides, for example, dicumylperoxide, di-tert-butyl peroxide, and/or di-tert-butyl cumyl peroxide preferably in amounts of about 1.4 to about 2.2% by weight. They can also be radiation cross-linked using gamma-radiation, for example, CO^{60} or high energy electron beam radiation in air or under nitrogen. Co-agents which enhance radiation cross-linking and enable a lower radiation dose to be used can also be added either during or subsequent to the mixing step preferably in an amount of about 1 to about 3% by weight. Examples of such co-agents include unsaturated allylic compounds, triallylcyanurate, acrylic compounds and acrylate or polyacrylate compounds. Protection against radiation damage to the (co)polymer can also be achieved by the addition preferably of up to about 2% by weight of radiation protectors such as trimethyl quinoline polymers or oligomers, for example, Age Rite Resin D. The (co)polymer and/or composition may also be cross-linked after grafting the (co)polymer or composition with an organic silane with the aid of a free radical initiator. Catalysts for cross-linking include DBTDL (di-butyl-tin-dilaurate) or dioctyl-tin-dilaurate (DOTDL).

The cross-linking of the peroxide cross-linkable composition or the resulting products may be conducted in steam or nitrogen under pressure at elevated temperatures, higher than the decomposition temperatures of the peroxides used to form free radicals. Radiation cross-linking is carried out at room temperatures. Silane cross-linking is carried out in the presence of water, steam or moisture at ambient or preferably at higher temperatures of up to about 90 to about 100°C. The speed and the duration of the cross-linking will depend on the type of (co)polymer and nanofiller used in the composition and the thickness of the composition.

It will be appreciated that one or more additives known in the art of polymer processing can also be included in the composition and added at any stage of the process. Suitable additives include antioxidants, for example, phenolic antioxidants such as SANTONOX R marketed by Monsanto and IRGANOX 1010 which is pentaerythritol tetrakis (3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate or IRGANOX 1035 which is octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate marketed by Ciba-Geigy or aminic antioxidants such as Vulcanox HS and Flectol H which are polymerised 2,2,4-trimethyl-1,2-dihydroquinoline; metal deactivators and/or copper inhibitors, for example, hydrazides such as oxalic acid benzoyl hydrazide (OABH) or Irganox 1024 which is 2,3-bis-((3-(3,5-di-tert-butyl-4-hydroxyphenyl) proponyl))propiono hydrazide; UV absorbers, for example Tinuvin or HALS type UV absorbers; foaming or blowing agents which may be either endothermic or exothermic for example, p.p-oxybis benzene-sulfonyl-hydrazide, azo-iso-butyro-nitrile and azodicarbonamide; processing and/or thermal stabilisers, for example tris (2,4-ditert-butylphenyl) phosphite (phosphite based), pentaerythritol tetrakis (3-(3,5-di-tertbutyl-4-hydroxyphenyl)propionate), octadecyl-3(3,5-di-tert-butyl-4-hydroxyphenyl)propionate, 3,3′,3′,5,5′,5′-hexa-tert-butyl-a,a′,a′-(mesitylene-2,4,6-triyl)tri-p-cresol

(phenolic based) and dioctadecyl-3,3'-thiodipropionate (thioester based); pigments, for example, inorganic pigments such as titanium dioxide and carbon black and organic pigments; flame retardants, for example, borates and metaborates such as zinc borate or metaborate, glass beads or particles, silica, silicon dioxide, compounds of silicon dioxide with other metal oxides; extenders, plasticisers or softeners, for example, polymeric plasticisers, phthalates such as dioctylphthalate, dioctylsebacate or dioctyladipate or mineral oils such as napthenic, paraffinic or aromatic oils.

The (co)polymers are preferably granulated, pelletised, powderised, cut and/or diced. The (co)polymer and the nanofiller can then be pre-mixed or added simultaneously, sequentially and/or separately to any suitable known apparatus, such as roll mills, internal mixers or continuous mixers for example twin screw mixers. It will also be understood that the known fillers and/or additives can be added simultaneously, sequentially and/or separately at any stage of the processing.

The nanofiller or composition may be exfoliated and/or delaminated using any suitable known technique such as high shear processing, for example, in the mixing apparatus referred to above. In variations of processes (a) to (c) defined above, a further exfoliation and/or delamination step may be performed.

In another embodiment of the processes of the invention, the (co)polymer, nanofiller and/or other additives are advantageously dry or dried in a separate step prior to processing involving hot air or dessicated hot air, in particular when silane grafting is used.

The composition of the invention can be formed by any suitable known process including moulding, such as injection moulding, blow moulding or compression moulding; pressing; vacuum forming; extrusion such as co extrusion, tandem extrusion or lamination with other layers for example polymeric layers; calendering and heat shrinking.

The heat shrinking process involves heating and stretching the composition and then cooling the composition in its stretched state. When the heat shrinkable articles are re-heated to temperatures above the crystalline melting point, they display shape memory properties, that is, they retain their original shape and size.

The composition of the present invention is either cross-linkable in the form of granules, pre-mixes or mixes, pellets, tapes or profile or intermediary, semi-fabricated articles or cross-linked in the form of intermediary, semi-fabricated or final articles. Examples of articles include profiles, tubes, pipes, films, sheet, tiles, floor coverings, containers and packaging for food.

The compositions of the present invention possess advantageous properties including high modulus and strength, increased barrier properties such as reduced penetration, permeation and/or lower diffusion of chemical solvents, oils and gases, reduced swelling, high heat distortion temperatures, increased dimensional stability, no melting, improved flame retardancy, lower specific gravity/density. These properties exist in particular at high temperatures.

Examples of applications of the composition include:

Medical: protective gear and clothing, medicine containers;

Defence applications and work protection: protection against external chemicals;

Transport: land, vehicles, trains, subways, sea, ships, air;

Construction: high rise, towers, installations and rooms with electronics, switches, computers, offices, public areas, theatres, cinemas, malls, stations, airports, telecom installations, storage;

Agriculture; and

Food: packaging of consumables, protecting food in laminated films.

EXAMPLES

The invention will now be described with reference to the following non-limiting examples.

The compositions of the examples were prepared using continuous co-rotating twin-screw mixers of the Werner & Pfleiderer ZSK type. Comparative examples 1 and 2 and Examples 1 to 3 on a ZSK-53 with the co-rotating screws of 53 mm diameter each with screw speeds of around about 200 rpm and feeding of about 50 kg/hour. Examples 4 to 8 were made on a ZSK-120 with the co-rotating screws of 120 mm diameter, with a range of about 150 to 180 rpm and a feeding rate of up to about 400 kg/hour.

The temperature was in the range of 190-220°C for LLDPE and 200-250°C for HDPE.

Comparative examples 1 and 2 and Examples 1 to 3 were mixed, grafted and exfoliated in one step, with the exception of some pre-mixed ingredients. Examples 4 to 8 were mixed and grafted in a first step and the nano-filler was then added and exfoliated in a second step. Some of the additives and nanofillers were added as pre-mixed or as a master-batch or concentrate.

A nitrogen blanket was used in each example (i.e. the feeding zone or zones were under nitrogen atmosphere for safety reasons and also for more efficient use of the peroxide radical initiator).

The compositions were granulated or pelletised directly at the exit of the ZSK mixers. Packaging was in metal lined bags of various size.

The silane grafted material was mixed with 4% of a catalyst masterbatch of e.g. DLDTP which is an accelerator directly prior to the formation of a product e.g. extrusion forming to tapes or to injection moulding for test plaques or prior to extrusion or extrusion of larger items or blow forming. These were then cross-linked in hot water, at temperatures of 90 to 110°C for 1 to 2 hours, depending of the thickness of the sample.

The percentage figures of the additives in the examples have been rounded to the first decimal after the point.

Comparative Example 1

HDPE MFI 0.2, granules (Type GM7655)....83.5%
HDPE MFI 25, powder (" GA 7260H)..14.7%
Silox VS 911..............................1.2%
Stabiliser Irganox 168 FF.................0.2%
Anox 20...................................0.4%

Hot Set Test (at 200°C):
Elongation under load.....................37%
residual elongation relaxed, no load.......0%

Comparative Example 2

LLDPE MFI 2.5 Alkatuff 425 gran........78.7%
LLDPE MFI 20 Ladene powder.............13.2%
Silox VS 924..............................1.4%
Irganox B 900.............................0.2%

Hot Set Test (at 200°C):
with load.................................270%*
without load..............................10%
* requires more Silox addition to 1.6%

Example 1

LLDPE Alkatuff 425 gran.................73.8%
LLDPE Ladene powder.....................18.4%
Silox VS 924..............................1.6%
MAH-HDPE Fusabond MB 100D.................1.0%

Irganox B 900..............................0.2%
Nanofiller Tixogel MP 100................5.0%

Hot Set Test (at 200°C):
under load.................................77%
without load................................0%

Example 2

LLDPE Alkatuff 425 gran....................74%
LLDPE Ladene powder......................18.5%
Silox VS 924..............................1.4%
MAH-HDPE Fusabond MB 100 D................1.0%
Irganox B 900.............................0.1%
Nanofiller Tixogel MP 100.................5.0%

Hot Set Test (at 200°C):
under load................................100%
without load................................0%

Example 3

LLDPE Alkatuff 425 gran..................79.3%
LLDPE Ladene powder......................14.0%
MAH-HDPE Fusabond MB 100 D................1.0%
Irgtanox 168 FF...........................0.2%
Anox 20...................................0.4%
Nanofiller Tixogel MP 100.................5.1%

Example 4

HDPE HMW Lupolen 2461A 88.1%

Stabiliser mix:
Irgafos 168FF+Anox20+Ca-Stearate(0.2+0.2+0.5)...0.9%
Fusabond MB 100 D...............................1.0%
Tixogel/Lupolen mix: Lupolen 2461A...........7.0%
Nanofiller Tixogel MP100........................3.0%

Example 5

HDPE GM 7655 gran....................................82.0%
Lupolen 4261 powder*(pre-mixed with following Silox).13.4%
Silox VS 911*(pre-mixed with above Lupolen powder)....1.6%
Process aid (masterbatch/concentrate).................1.0%
Stabiliser/Antioxidant mix (masterbatch/concentrate)..3.0%

The Pre-mix of Lupolen and Silox has been high speed pre-mixed and added via a separate feeder.

Hot Set Test (at 200°C):
Elongation with load...........................160%
Elongation relaxed (after removing load).........7%

Example 6

HDPE GM 7655 gran*..........................73.0%
HDPE GA 7260H powder*.......................11.9%
Silox VS 911*................................1.4%
Antioxidant* (masterbatch/concentrate).......1.8%
Process aid* (masterbatch/concentrate).......0.9%
MAH-HDPE Fusabond 100 D......................1.0%
Composite mix**(including nano-filler)......10.0%
(above 10% comp.mix**consisting of:
Nanofiller Tixogel MP........................3.0%
Irgafos 168 FF...............................0.2%
Calcium stearate.............................0.5%
HDPE, MFI 20, MG 20224 powder................0.9%

Lupolen 4261A....................................2.1%
Compound Example 5...............................3.1%
Anox 20..0.2%
Subtotal of composite mix**.....................10.0%

* The components were pre-mixed & grafted separately.

Hot Set Test (at 200°C):
elongation under load..........................173%
elongation with load removed (relaxed).........7%

Example 7

HDPE MFI 10, HD1090 gran......................88.3%
HDPE MFI 20, Ladene powder.....................9.7%
Silox VS 924...................................1.6%
Irgafos 168 FF.................................0.2%
Anox 20..0.2%

(after grafting prior to forming catalyst masterbatch/concentrate was added 5% in most other examples 4% cat.MB was added)

Hot Set Test (at 200°C)
load 250%
relaxed 16%

Example 8

Compound Example 7:...........................86.9%
HDPE MFI 20, Ladene powder.....................8.7%
Nanofiller Tixogel MP100.......................3.0%
Stabiliser mix.................................1.4%

Hot Set Test (at 200°C):

load........................167%
relaxed 13%

Many modifications may be made to the preferred embodiment as described above without departing from the spirit and scope of the present invention.

Dated this 28th day of March 2002
COMPCO PTY LTD
By their Patent Attorneys
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Fellows Institute of Patent and
Trade Mark Attorneys of Australia